



## Development of a personal aerosol sampler for monitoring the particle-vapour fractionation of SVOCs in workplaces

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Keywords:	gas-particle partitioning, semi-volatiles, personal sampling, denuder, occupational health

## Short Communication

# Development of a personal aerosol sampler for monitoring the particle-vapour fractionation of SVOCs in workplaces†

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† Online Supplementary Material available

**Keywords:** Gas-particle partitioning; semi-volatile organic compound (SVOC); personal sampling; denuder.

## Abstract

Semi-volatile organic compounds (SVOCs), partitioned between particulates and vapours of an aerosol, require special attention. The toxicological effects caused by the inhalation of such aerosols may depend on the concentration and in which phase the organic compounds are found. A personal denuder-gas-particle separation (D-GPS) aerosol sampler was developed to provide information about the partitioning of aerosols from organic compounds. The sampler was tested in a series of controlled laboratory experiments, which confirmed the capability and accuracy of the sampler to measure gas-particle mixtures. An average difference of  $14.8 \pm 4.8$  % was found between sampler and reference laboratory instruments. The obtained results showed that our sampler enables a more accurate measurement of the SVOC aerosols' gas-particle fractionation, compared to that of conventional samplers.

## Introduction

Aerosols from semi-volatile organic compounds (SVOCs) such as polycyclic aromatic hydrocarbons (PAHs), metalworking fluids (MWFs), phthalates, etc. represent an important challenge to industrial hygienists due to toxicological as well as sampling issues. Mainly problematic is the sampling of hazardous SVOCs that are present as vapours and particulates in the workplace (Perez and Soderholm, 1991). Such compounds can be released into the air due to mechanical, thermal or combustion processes. The partitioning of SVOC aerosols is highly influenced by environmental factors, which lead to a continuous mass transfer taking place between the aerosol's particles and vapours (Dragan et al., 2014; Dragan et al., 2017). Previous studies have highlighted the potential measurement bias related to gas-particle partitioning, which is affected by environmental conditions and by the sampling method used. Volckens and Leith (Volckens and Leith, 2003) developed generalized equations to predict the sampling bias for filter-adsorbent, filter-filter-adsorbent and denuder-filter-adsorbent methods. Other authors (Kaupp and Umlauf, 1992) found substantial differences between sampling methods when using filters, impactors and electrostatic precipitators to measure the partitioning of semi-volatile PAHs and pesticides. Koutrakis et al. and Tsai et al. (Koutrakis et al., 1989; Tsai et al., 2001) studied the concept of coated denuders to trap gas phase inorganic acids and integrated the denuder into a personal sampler in order to measure human exposures to acid aerosols with better accuracy.

MWF mists represent one example of aerosols containing SVOCs emitted in workplaces. Workplace exposure to MWF aerosols can lead to a series of occupational respiratory diseases and even to lung cancer (Robertson et al., 1988; Kennedy et al., 1989; Robins et al., 1997; Calvert et al., 1998; Gauthier, 2003; Cohen and White, 2006; Lillienberg et al., 2010). Therefore, occupational monitoring should be performed regularly, in order to ensure workers are not at risk. In addition to the concentration workers are exposed to, the

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3 physical state of the inhaled compounds might influence the adverse health effects triggered  
4 by the inhaled aerosols. Compared to particles, gases and vapours are more likely to be  
5 exhaled (Gunderson and Anderson, 1987). The particle deposition in the lungs is a function  
6 of aerodynamic diameter, whereas gas deposition is a function of tissue solubility (Volckens  
7 and Leith, 2003; Heyder, 2004). For chemical agents that are not readily soluble in the tissue  
8 of the respiratory tract, the main uptake mechanism will be through deposition of particles.  
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17 Occupational monitoring of SVOC aerosols is usually performed using personal  
18 samplers with integrated filter cassettes and/or adsorber cartridges (Breuer, 1999; Simpson,  
19 2003; Breuer et al., 2011; Galea et al., 2011; Breuer et al., 2015; Kirkhus et al., 2015; Sutter  
20 et al., 2018). However, SVOCs collected on filters are in constant exchange with the air  
21 pumped through the filter and are likely to evaporate therefrom (Riss et al., 1999; Raynor et  
22 al., 2000; Sutter et al., 2010; Breuer et al., 2015; Dragan et al., 2015; Dragan et al., 2017).  
23 Consequently, a differentiation between vapours and particles is impossible, since the filter-  
24 adsorber approach only provides a consistent way to measure the sum of both phases. If a  
25 sampler fails to collect both phases of an aerosol separately, it can lead to an underestimation  
26 of the exposure to the particulate phase and an overestimation of the exposure to the vapour  
27 phase. This in turn can lead to a misinterpretation of the exposure data. Therefore, a further  
28 partitioning tool is required to achieve a more accurate measurement of both aerosol phases.  
29 This tool should also be simple enough to potentially be integrated into the routine analysis  
30 carried out by occupational hygienists. Furthermore, the backpressure generated by the  
31 additional partitioning tool should be minimal, so that it can still be operated with the  
32 available personal sampling pumps. The new assembly should be light, robust, reliable and  
33 not too cumbersome to be worn by workers for several hours.  
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### ***Considerations towards the sampler development***

The denuder-gas-particle separation (D-GPS) sampler (Figure 1) presented here was inspired by the GGP-Mini (*Gesamtstaub-Gas-Probenahme* – inhalable dust/gas sampling) personal sampler (Breuer et al., 2015) and by the silicone rubber trap denuders (Forbes et al., 2012; Geldenhuys et al., 2015; Kohlmeier et al., 2017; Munyeza et al., 2018). The samplers conical shape inlet is not size-selective and was designed similar to that of the GGP-Mini. The D-GPS sampler combines a denuder as a partitioning tool for vapours with a filter followed by a backup adsorber tube for the particle phase. Adsorber tubes can be used to trap the particle mass that evaporates from the preceding filter (*blow-off* effect).

The silicone rubber trap denuders considered for this study were designed to trap vapours of SVOC while allowing particles to pass through them. The denuders were extensively characterised in a series of laboratory particle number transmission tests, as well as vapour collection tests (Forbes and Rohwer, 2009; Forbes et al., 2012; Forbes et al., 2013; Kohlmeier et al., 2017; Munyeza et al., 2019). Within the particle number transmission tests it was shown that ambient particles (size distribution ranging from 40 to 400 nm) can pass through the denuder with an efficiency of 95 % (5 % particle losses inside the denuder) (Munyeza et al., 2019). 91-100 % of polystyrene latex (PSL) test particles in the range of 0.3 to 2  $\mu\text{m}$  passed vertically orientated denuders (i.e. 4 % losses at 0.5  $\mu\text{m}$ , 4 % at 1  $\mu\text{m}$ , 9 % losses at 2  $\mu\text{m}$ ) at a flow rate of 0.5 l min<sup>-1</sup> (Kohlmeier et al., 2017). The denuders' ability to trap vapours was demonstrated in a series of experiments with three model SVOC substances of different polarity, as well as with a theoretical prediction model (Kohlmeier et al., 2017). In this study, a maximal experimental vapour collection efficiency of 97 % was observed for *n*-hexadecane. The previously characterised denuders were therefore found suitable to trap vapours and were integrated into the D-GPS sampler. The focus of this study was to design

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3 and validate the personal sampling device D-GPS, that is able to separately trap the vapour  
4 and particle phases of SVOC aerosols.  
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## 11 **Materials and methods**

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15 A series of laboratory tests with generated *n*-hexadecane aerosols were conducted to  
16 demonstrate the accuracy in measuring the gas-particle fractionation of the new D-GPS  
17 sampler. *n*-Hexadecane test aerosols with six different particle to gas phase mass  
18 concentration ratios with particle sizes between 0.8 and 1.3  $\mu\text{m}$  were generated to compare  
19 *off-line* D-GPS sampling with *on-line* reference series measurements taken in parallel. In this  
20 way a validation of the new sampler could be achieved. The *off-line* gas phase values were  
21 obtained after thermal desorption of the D-GPS samplers' denuders. The *off-line* particle  
22 phase masses were calculated by summing up the *n*-hexadecane masses obtained after  
23 thermal desorption of the personal samplers' filters and adsorbers.  
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37 The detailed description of the D-GPS sampler and the laboratory characterisation  
38 experiments can be found in the Online Supplementary Material, available at Annals of Work  
39 Exposures and Health online.  
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## 48 **Results and discussion**

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50 The D-GPS samplers' denuders and adsorbers contained detectable amounts of *n*-hexadecane  
51 for all tests. On the other hand, the quantity of *n*-hexadecane found on filters was  
52 systematically below the quantification limit. This was a consequence of the vapour being  
53 collected in the denuder, enhancing the evaporation rate of particles from the filters. The  
54 particle mass evaporated from the filters was trapped on the adsorbers. Compared to the FID  
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3 reference series measurement, the total mass of *n*-hexadecane collected and quantified with  
4 the D-GPS samplers led to an average recovery rate of 95.9 % and a standard deviation of  
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6 9 %. There were no substantial variations of the recovery rate throughout the measurements.  
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8 The high recovery rate confirmed that the aerosols trapped on the sampler can be effectively  
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10 analysed using thermal desorption.  
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15 The D-GPS sampler has successfully estimated the gas-particle fractionation of  
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17 *n*-hexadecane model aerosols (Figure 2 and Table S1, see Supplemental Material). The  
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19 difference in gas-particle fractionation, as measured by both methods was found to vary  
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21 between 8.1 and 21.4 %, with an average difference of  $14.8 \pm 4.8$  % regarding the particle  
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23 phase. The slight differences regarding the particle mass obtained with the D-GPS can be  
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25 partially attributed to impaction of particles inside the denuder. Nonetheless this slight  
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27 difference in particle-vapour fractionation is reasonable, given the complexity of sampling  
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29 and analysis of the samplers three substrates.  
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34 These results show that an accurate fractionation of the generated test aerosol is  
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36 possible and represent an improvement compared to results obtained with the conventional  
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38 filter-adsorber method. The fractionation of SVOC aerosols using conventional samplers can  
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40 be substantially affected by particle mass evaporation. This was demonstrated in a previous  
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42 study of Breuer and Dragan (Breuer et al., 2015) where conventional filter-adsorber samplers  
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44 showed that for a similar concentration of *n*-hexadecane aerosols to this study of  $10 \text{ mg m}^{-3}$ ,  
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46 no particle mass was found at all on the on filters of the GGP-mini off-line samplers. This  
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48 represented an underestimation of the particle mass of about 40 %, as approximately  $4 \text{ mg/m}^3$   
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50 of particle-bound hexadecane was measured in parallel by the reference method. In contrast,  
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52 the D-GPS sampler presented here only underestimated the particle phase on average by  
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54 14.8 %. The particle evaporation rate from filters of conventional samplers can be further  
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56 enhanced by higher temperatures. On the other hand, the aerosol fractionation measured with  
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3 the D-GPS will not be influenced by temperature. The present study demonstrates that the  
4 developed sampler can provide reliable information regarding gas-particle fractionation, with  
5 similar analyte recovery rates to conventional filter-adsorber samplers.  
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### 9 10 ***Study limitations***

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13 Only one compound class (linear alkanes) and one type (hexadecane) was studied here. It is  
14 therefore possible that the range of polarity and vapor pressure encountered in the field would  
15 lead to different breakthrough/adsorption capacities that have not been studied and reported  
16 here. The D-GPS sampler has not yet been tested or validated on a worker, which could lead  
17 to potential complications that are, as of yet, unknown. The particle penetration through the  
18 D-GPS has not been tested in the fully respirable, thoracic, or inhalable range. It may be that  
19 particles larger than 2  $\mu\text{m}$  experience greater losses through the denuder.  
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### 34 **Conclusion and perspectives**

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37 The D-GPS was tested under laboratory conditions with generated *n*-hexadecane aerosols of  
38 six different particle-vapour ratios. The sampler proved to accurately measure the vapour-  
39 particle fractionation of the aerosol, with an accuracy of  $14.8 \pm 4.8 \%$  and an average  
40 analytical recovery rate of 95.9 %. The sampler is lightweight, operated with battery-powered  
41 personal sampling pumps and all three sampling substrates were successfully analysed using  
42 thermal desorption - gas chromatography.  
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51 Since currently in Germany available personal aerosol samplers are only able to sample the  
52 “total SVOC aerosol mass”, samplers such as the D-GPS represent a new approach towards a  
53 simultaneous measurement of both aerosol phases. The described D-GPS sampler may enable  
54 a better interpretation of the exposure data with regard to the gas-particle distribution of  
55 SVOC aerosols. Furthermore, the D-GPS samplers can lead to a reconsideration of  
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3 occupational exposure limits (OELs), together with toxicological studies on different effects  
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5 caused by exposure to particulate and gaseous compounds.  
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## 10 **Acknowledgements**

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18 acknowledged.  
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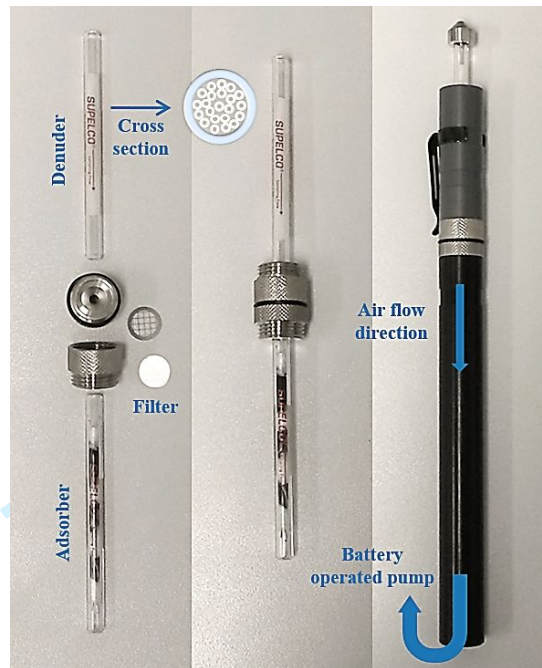
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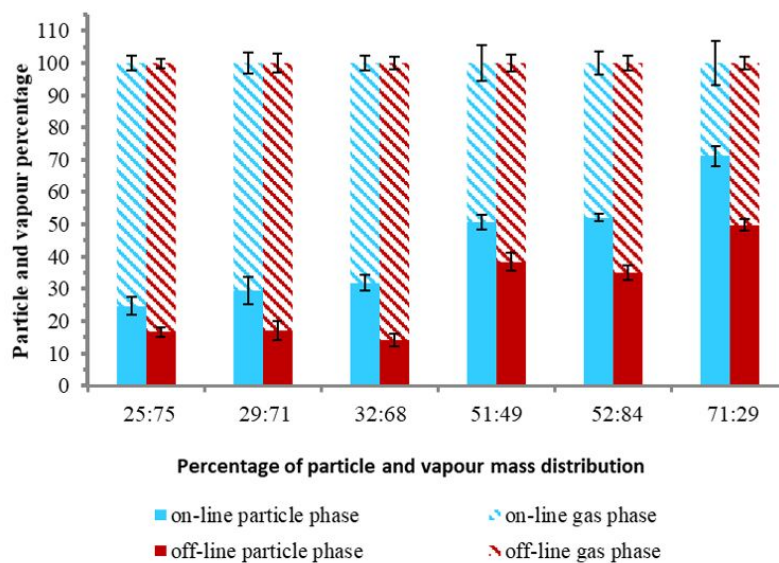
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## Figures



**Figure 1** Portable denuder-gas-particle separation (D-GPS) aerosol sampler disassembled and assembled with inlet and robust housing.



**Figure 2** Six comparison measurements carried out *on-* and *off-line* with six different particle to gas phase mass concentration ratios (abscissa) of an *n*-hexadecane aerosol as determined by the *on-line* technique. For each comparison measurement, the minimum number of replicates was 4 (*off-line* sampling) and 2 (*on-line* series measurements). Each error bar indicates one coefficient of variation (*on-line* series measurements) or the standard deviation (*off-line* measurements).

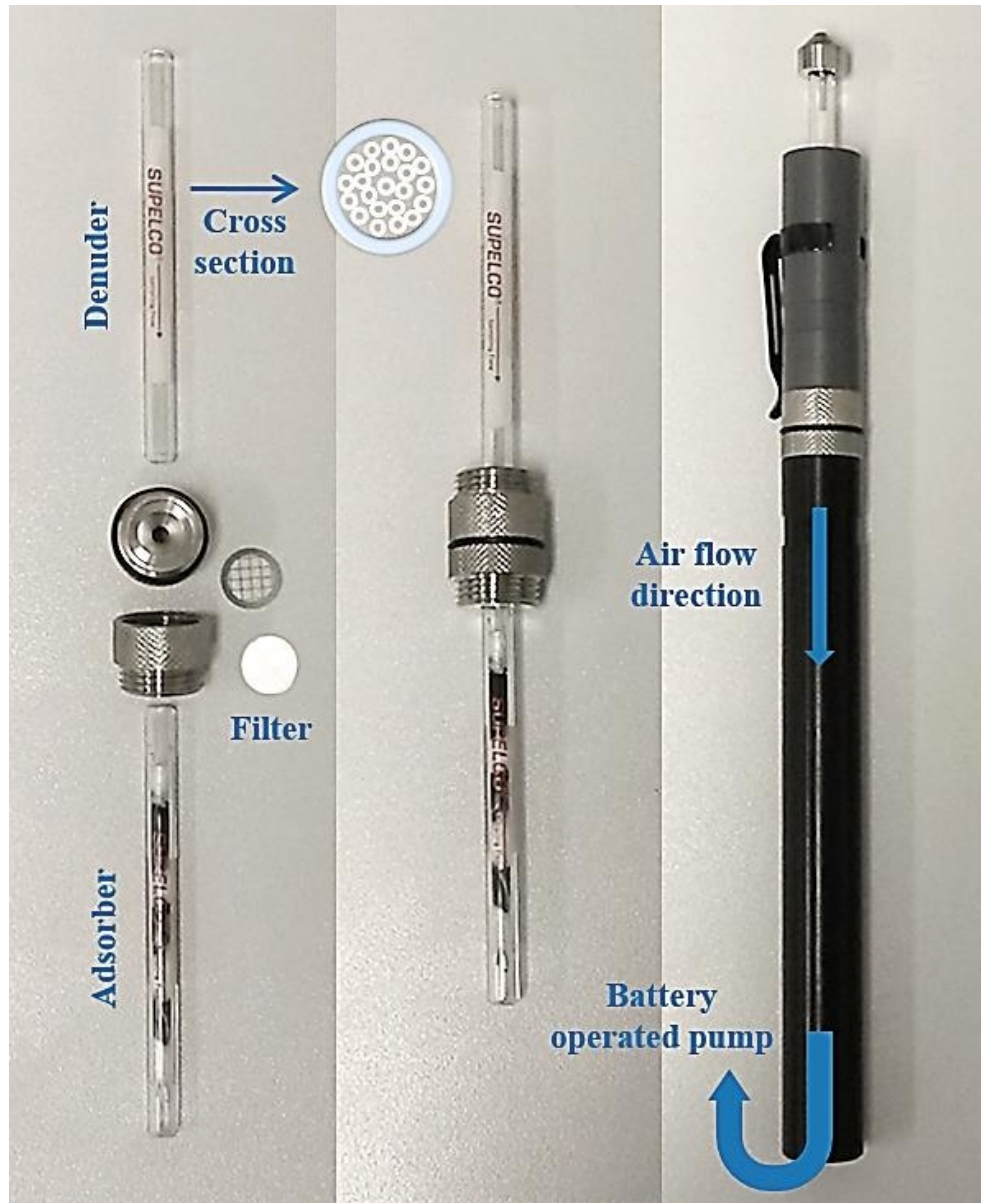
### **Conflict of interest declaration for publication**

Funding for this project was provided by German Social Accident Insurance (DGUV). The authors declare no conflict of interest relating to the material presented in this article. Its contents, including any opinions and/or conclusions expressed, are solely those of the authors.

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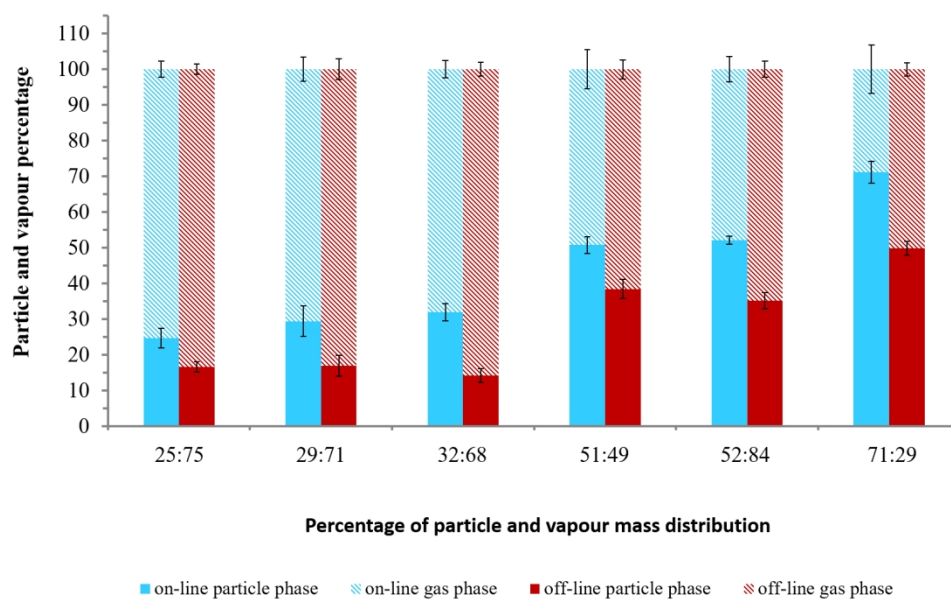
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Portable denuder-gas-particle separation (D-GPS) aerosol sampler disassembled and assembled with inlet and robust housing.

100x123mm (150 x 150 DPI)





Six comparison measurements carried out on- and off-line with six different particle to gas phase mass concentration ratios (abscissa) of an n hexadecane aerosol as determined by the on-line technique. For each comparison measurement, the minimum number of replicates was 4 (off-line sampling) and 2 (on-line series measurements). Each error bar indicates one coefficient of variation (on-line series measurements) or the standard deviation (off-line measurements).

146x94mm (300 x 300 DPI)